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Abstract

Processes for the Recovery and Purification of Fission Products from Irradiated Uranium

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Waste solutions from reprocessing plants may be treated in order to achieve one or more of the following objectives: -

1. Solutions still containing the bulk of the uranium: To separate most of the uranium from the fission products in order to achieve a high concentration factor for the final waste.

2. To separate scientifically or technically valuable and long-lived fission products, such as ^{137}Cs, ^{90}Sr, ^{144}Ce and ^{147}Pm, in a pure form.

3. To separate most of the long-lived nuclides ^{137}Cs and ^{90}Sr in order to considerably shorten the requested storage period of the high activity waste.

Among the various methods conceivable we have so far tried ion exchange using inorganic compounds, and also freezing out. These methods may, of course, also be combined with others, e.g. precipitation.

Inorganic Ion Exchangers

Zirconium phosphate and the hydrous oxides of zirconium and silicium (silica gel) have been judged as the inorganic ion exchangers of greatest promise. Zirconium phosphate is moderately acidic, the hydrous oxides only weakly. In fact the zirconium hydrous oxide is amphoteric, acting mainly as an anion exchanger when pH $<$ 5. The selectivity between ions of different charges generally increases with decreasing acidity. On the other hand, a certain

degree of sorption is reached at a lower pH value, the more acidic the gel which may be important for easily hydrolysed ions. For pH > 8, zirconium phosphate is heavily hydrolysed and thus not useful as an ion exchanger. Silica gel can be used up to a pH = 9, where dissolution becomes appreciable. The workable ranges of pH, as well as other properties of the investigated ion exchangers, thus supplement each other rather well.

With zirconium phosphate, Cs^{+} and Ce^{3+} have been isolated from Sr^{2+} as pure fractions, even in the presence of large amounts of Na^{+}. It may even be possible to isolate Cs^{+} from solutions containing UO_2^{2+} in rather high concentrations.

With silica gel Zr(IV) may be selectively sorbed directly from the strongly acidic waste. This cannot be achieved with zirconium phosphate, which reacts irreversibly with four-valent cations, on account of their great affinity to phosphate ions.

Separation of Uranium from Fission Products by Freezing

On partial freezing of a concentrated solution of uranyl nitrate containing a small amount of fission products, uranyl nitrate and ice crystallize leaving the fission products in the mother liquid. The phases may be separated by centrifugation, leaving the solid somewhat contaminated by mother liquid. By repeated partial freezing it should be possible to purify the uranyl nitrate further. The more important fission products have been tested and do not appear to co-crystallize with uranyl nitrate. The freezing out of uranyl nitrate and ice may also be performed in the presence of moderate concentrations of nitric acid, which reduces the solubility of the uranyl nitrate.

It thus seems physically possible to recover a considerable fraction of uranium in a rather pure form from a concentrated uranyl nitrate solution containing small amounts of fission products by repeated fractional freezing. Simultaneously the high activity waste is concentrated in a reduced liquid volume.